

Zeolite supported Pd electrocatalyst nano-particle characterization

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INTRODUCTION

Palladium (Pd) as one of platinum (Pt) group metals has attracted significant attentions since it has shown good activities for hydrogen reaction (HOR) and oxygen reduction reaction (ORR) in fuel cell application and high solubility and mobility as hydrogen storage material¹. Furthermore, due to considerably cheaper price than Pt, Pd is regarded as a very compatible substitute alternative to Pt once its electrocatalytic performance can be further enhanced in coherence to Pt catalysts. Over the years, extensive researches have been carried out to investigate Pd nano-structure and surface activity on carbon based Pd system, with few studies having focused on polymer materials like zeolite based Pd electrocatalyst. Hence, this paper contributes towards the study of laboratory made zeolite supported 1.5 wt% Pd nanoparticle structure and its resultant electrochemical performance for fuel cell application by using cyclic voltammetry (CV) and the Extended X-ray Adsorption Fine Structure (EXFAS).

EXPERIMENTAL/DATA ACQUISITION

The 1.5wt% Pd decorated Y-zeolite electrocatalyst was made using $(\text{NH}_3)_4\text{PdCl}_4$ in a neutral solution of PH⁷². The synthesis of Pd nanostructure on Y-zeolite was carried out using calcination and reduction procedures to remove Pd co-ordinate ligand at 350 °C in O₂ and reduced at 250°C, 360°C and 400°C, respectively in 5% H₂/95%N₂ gas mixture (therefore denoted as 15Pdanc25r36, 15Pdanc35r36, and 15Pdancr4). The electrode was made of samples with 1.5 wt% Pd loading on zeolite and XC-72R carbon powder (untreated) mixed with 15 wt% Nafion® solution as a binder for CV measurement. A pellet made by mixture of 1.5 wt% Pd zeolite samples and boron nitride powder was used for en-situ EXAFS measurement using Synchrotron Radiation Source (SRS) at STFC Daresbury national Laboratory, UK. The data analysis was carried out using in-house software EXCURV 98.

RESULTS AND DISCUSSION

The CV measurement presented in figure 1(a) shows a similar hydride region including hydrogen desorption, re-oxidation, adsorption and evaluation peaks for Pd zeolite catalyst of 15Pdancr4 and commercial Pd catalyst with 1.5 wt% Pd loading on XC-72R carbon (denoted as 15PdXC-72R). The single hydrogen re-oxidation peak observed for Pd zeolite catalysts of 15Pdanc25r36 and 15Pdanc35r36 indicates very similar energy levels of hydrogen desorption and re-oxidation on Pd surface. Figure 1(b) depicts a typical EXAFS Chi data fitting for catalyst of 15Pdancr4. The particle size made of sample

15Pdancr4 was found the largest, followed by that of 15Pdanc35r36 and 15Pdanc25r36, respectively. The Pd particle size was predicted at a range of 0.82 – 0.93 (nm). Conclusively, large particle size offers better electrochemical perform, supported by an evidence of current change at hydride region for catalysts in an order of 15Pdancr4 > 15Pdanc35r36 > 15Pdanc25r36. It was found that the 15PdXC-72R has no clear advantage over the Pd zeolite catalysts. This tendency is contradicting to that previously observed for 1.5 wt% Pt zeolite catalysts², implying the metal location on zeolite has to be taken into account. Also, Pd presents strong mobility than Pt, due to high charger density. Pd particles are more favorable on zeolite supercages or external surface at high calcinations and reduction temperature, whilst the most of Pd particles were entrapped on zeolite sodalite or hexagonal cages at low temperature. The hydrogen adsorption on Pd surface might experience an up-hill diffusion process and surface conductance via β -PdH or α -PdH hydride phases³.

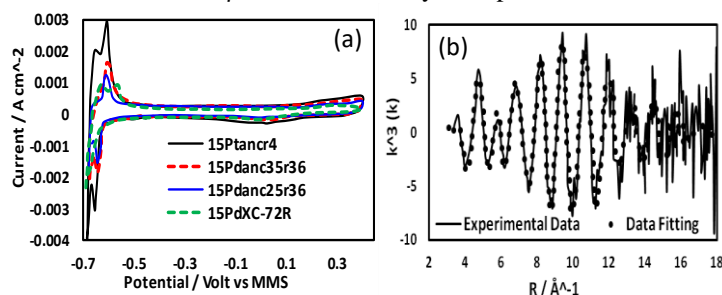


Fig. 1 Pd decorated zeolite electrocatalyst. (a) The CV comparison for samples of 15Ptancr4, 15Ptanc25r36, 15Ptanc35r36 and 15PtXC-72R, respectively. (b) The EXAFS Chi data fitting for sample 15Ptancr4.

CONCLUSION

Pd nano particle location on zeolite is important for electrochemical performance. The hydrogen up-hill diffusion process and surface conductance pathways might be equally contributed for electrochemical reaction that are taken place on Pd surface while forming β -PdH and α -PdH hydride phases. The future study will be focused on small size organic species oxidation on Pd decorated zeolite electrocatalyst.

REFERENCES

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